Characterization of Ceramics and Intermetallics Fabricated by Self-Propagating High-Temperature Synthesis

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CHARACTERIZATION OF CERAMICS AND INTERMETALLICS FABRICATED BY

SELF-PROPAGATING HIGH-TEMPERATURE SYNTHESIS

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SUMMARY

This report summarizes three efforts aimed at investigating the process of self-propagating high temperature synthesis (SHS) for the fabrication of structural ceramics and intermetallics. Of special interest was the influence of processing variables such as exothermic dopants, gravity, and green state morphology in materials produced by SHS.

In the first effort directed toward the fabrication of SiC, exothermic dopants of yttrium and zirconium were added to SiO_2 or SiO_2+NiO plus carbon powder mix and processed by SHS. This approach was determined to be unsuccessful since it did not produce the desired product of crystalline SiC.

In the second effort, the influence of gravity was investigated by examining Ni-Al microstructures which were produced by SHS combustion waves traveling with and opposite the gravity direction. Although final composition and total porosities of the combusted Ni-Al compounds were found to be gravity independent, it was observed that larger pores were created in those specimens which were combusted opposite to the gravity force direction.

Finally, it was found that green microstructure has a significant effect on the appearance of the combusted piece. Severe pressing laminations were observed to arrest the combustion front for TiC samples.

INTRODUCTION

Self-propagating high-temperature synthesis, or SHS, has potential to be a new synthesis method for many ceramics and alloys which are of interest to NASA. The advantages of the SHS method include high purity end-products as the high temperatures involved vaporize volatile impurities. Also, very short process times, on the order of seconds or less, are needed as opposed to hours or days for normal sintering. Finally, SHS does not require a high temperature furnace for processing.

For SHS, loose powder of two or more elements or compounds are mixed and may be formed into a shape (although attempts have also been made to create nitrides by mixing a solid with a gas or liquid (refs. 1 and 2). This shape or loose powder mixture is ignited by a thermal source such as an electrical heating wire. Ignition results in an adiabatic chemical reaction between the elements and/or compounds. The heat release causes ignition of adjacent powder resulting in a combustion wave passing through the powder or shape leaving behind a different compound or alloy than the starting materials. The

adiabatic temperature of the reaction, T_{ad}, can be determined from the following equation

$$\Delta H_{To} = \int_{T_o}^{T_m} C_p(s) + \Delta H_f + \int_{T_m}^{T_{ad}} C_{p(1)} \, dT$$

where ΔH_{To} is the enthalpy at T_o, $C_p(s)$ and $C_{p(1)}$ are the heat capacities of the product in its solid and liquid states, and ΔH_f is the enthalpy of fusion at the melting point of the product, T_m. This generalized equation is for the case when the final product is completely in the liquid state. In many commonly investigated combustion systems such as TiC and TiB_2, only partial melting occurs, where $T_m = T_{ad}$. In this case, the second term of the above equation is replaced with $V \Delta H_f$, where V is the fraction of the product in the liquid state. It has been empirically found that a T_{ad} of 2000 °C is necessary to easily ignite a specimen (ref. 1). This has made it difficult to produce materials by the SHS process from weakly exothermic systems such as Si + C.

In 1984, NASA Lewis Research Center and Los Alamos National Laboratory (LANL) undertook a joint project entitled "Solid State Combustion Synthesis of Ceramics and Alloys in Reduced Gravity". This project, proposed by LANL, primarily involved modeling efforts and sample preparation on the part of LANL with NASA Lewis analyzing microstructures of specimens provided by LANL. This report will summarize NASA Lewis efforts on this project. LANL efforts are reported elsewhere (ref. 3). The NASA Lewis effort fell into three categories. The first effort was analysis of potential new ceramic materials developed by LANL. Several systems were attempted which were expected to form SiC based compositions.

SiC is an important candidate for high temperature applications and so has been studied at Lewis for a number of years. As Si+C→SiC is only a weakly exothermic reaction (T_{ad} = 1502 °C), it is difficult to initiate ignition. However, there have been several successful attempts to fabricate SiC (refs. 4 to 6). One method has been to utilize high pressure to increase the ignition area in order to enhance the reaction (ref. 4). Another method has been to use nonstandard ignition methods such as those reported by Yamada et al. (ref. 5) and Wei (ref. 6) which both produced powders. These efforts involved attempts to make pure SiC. In the current study, it was attempted to fabricate a composite material based on SiC with a second phase. The second phase was one with a high heat of formation which was hoped would act as an energy source for a standard ignition pressureless reaction. Avoidance of high pressure methods of fabrication was considered useful as expulsion of adsorbed gases from the powder surfaces while under pressure is a difficult problem to deal with experimentally. Also, a second phase can act as a strengthening mechanism (ref. 7) for structural ceramics. SiC has been extensively investigated for the past 20 years for structural applications. The microstructures of the resultant materials are discussed in the first section of the Results portion of this report.

The second area involving NASA Lewis was analysis of several specimens which were ignited from either the top or bottom of Ni-Al samples. This work was to support LANL modeling efforts by S. Valone on the effect of gravity. Ni-Al compositions were chosen because $T_{ad} > T_m$, so melting would occur

during combustion and thus increase the likelihood of observing an effect of gravity via bulk flow. One would not expect to see an effect of gravity in a solid state reaction where $T_{ad} < T_m$, or perhaps not even in a reaction with a small amount of liquid, where $T_{ad} = T_m$, due to the influence of capillary forces. The Ni-Al system has recently been extensively investigated by Philpot, Munir and Holt (ref. 8). Microstructural examination was to search for effects of gravity on the resultant microstructure.

The last category was a NASA Lewis initiated effort to better understand the effect of green microstructure on the combusted microstructure of TiC. This work looked specifically at the effect of pressing laminations. Other aspects of green processing effects on SHS have been studied by others (ref. 9).

EXPERIMENTAL PROCEDURE

The bulk of the specimens in this study were prepared at LANL by standard SHS techniques. These specimens included the samples designed to produce SiC plus a second phase as well as the Ni-Al samples. Reagent grade raw materials were mixed together by hand, pressed into pellets which were right circular cylinders of 2 cm in height by 1 cm in diameter. These pellets were then ignited by a tungsten wire positioned above the sample so that the combustion wave traveled along the longest dimension of the pellet. The pellets sat on a graphite strip. Combustion occurred in at a slightly negative pressure with about one ppm O_2 present. The power source had a 500 A capacity. Ignition conditions and temperatures are not available. Samples dimensions were obtained and the samples were then sent to NASA Lewis. Once at Lewis, the specimens were sectioned longitudinally through the center of the samples. X-ray diffraction analysis was performed on powder which was ground from one of these sectioned halves. The other half was mounted and polished for metallography and microprobe analysis.

Specimens which were prepared at Lewis were TiC and were treated in a similar manner to the LANL specimens. However, raw powders were dry mixed or wet mixed in a mill and will be discussed further in a subsequent section. Combustion was carried out in a flowing argon atmosphere, but ignition was initiated on a graphite strip rather than by a tungsten wire. The power supply was a 500 A source. Ignition was carried out with 5.5 V and 300 A. The power was applied for about 1 sec before ignition began. Power was shut off as soon as ignition occurred.
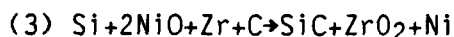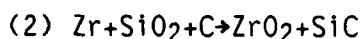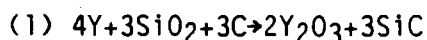
RESULTS

Ceramic Synthesis

In addition to examination of gravity effects on the SHS process, the LANL research included investigation of the synthesis of new ceramics of potential interest as structural materials. In conjunction with this interest, Dr. Robert Behrens and Henry Brush made a presentation (ref. 10) at the DARPA/Army Symposium on SHS involving calculating equilibrium phase diagrams and equilibrium vapor compositions for the Y-Si-C-O and Zr-Si-C-O systems. As SiC is an important structural ceramic, it was suggested by Dr. James Smialek of NASA Lewis that SiC could be prepared with Y_2O_3 or ZrO_2 as a

second phase, both of which are known to have high heats of formation in ceramic systems. Also, ZrO_2 is known to be an effective toughening agent in ceramic systems (ref. 7). A highly exothermic reaction to create a second phase was necessary to evolve a large amount of heat as the $Si + C \rightarrow SiC$ reaction alone is only weakly exothermic. In support of this work, samples were given to NASA Lewis for analysis. The samples were expected to represent the following reactions:

(1) $4Y+3SiO_2+3C \rightarrow 2Y_2O_3+3SiC$

(2) $Zr+SiO_2+C \rightarrow ZrO_2+SiC$

(3) $Si+2NiO+Zr+C \rightarrow SiC+ZrO_2+Ni$

In these reactions, SiO_2 and NiO act as oxygen sources with NiO a less stable source than SiO_2. SiO_2 is also a Si source for SiC. For each reaction, C was added as a carbon source for SiC.

Each sample was first analyzed by x-ray powder diffraction to determine the phases present. They were also examined by scanning electron microscope (SEM) and by electron microprobe.

The x-ray results for each system are found in table I. These results indicate that crystalline SiC was not formed in any system. Reaction 1 and 3 appeared to be the most promising with at least one of the proposed end-products occurring in each case, Y_2O_3 and ZrO_2, respectively. However, additional phases were also present. YSi_2 was found in the case of the Y-Si-C-O system in reaction 1 and ZrC in the Ni-Zr-Si-C-O system in reaction 3. Reaction 2 in the Zr-Si-C-O system produced only Zr and C according to x-ray results. However, x-ray data cannot be interpreted to suggest SiC does not exist. Poorly crystallized or amorphous phases could be present.

By optical microscopy, all samples appeared to be very inhomogeneous. Extensive areas of large porosity were found adjacent to dense areas. Within dense regions, microstructures were multiphase and inhomogeneous. These structures are shown in figure 1. Elemental mapping by electron microprobe was performed to learn more about these phases and to see if inferences could be drawn regarding the presence of poorly crystallized or amorphous phases.

Figure 2(a) contains mapping results for the Y-Si-C-O system produced by reaction 1. This is a representative microstructure within the region shown in figure 1. The first photo is a BSE photo showing an interesting micro-structure and the degree of inhomogeneity present in this sample. Mapping identified the white area in this photo as localized Ta impurities. The dense center region is a Si-Y-C phase. The center phase is surrounded by a region with the appearance of a eutectic composition of Si-Y-C. Possibly this phase is the one identified as YSi_2 by powder diffraction. On the outer edge of this phase are discrete, angular black areas, totally deficient in O and Y, but clearly containing Si. Carbon may be there as well. On the outer areas of the photograph, the matrix is composed of Y and O with C present as well but at a reduced concentration. This region may be composed primarily of the phase identified as Y_2O_3 by x-ray diffraction. Figure 2(b) has the mapping results from a different area of the same sample. Ta contamination exists in this area as well. In the center of the BSE photo is a black angular phase which is composed of Si and C. This is probably the same angular

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black phase present in the previous figure. Additionally, the outer matrix and inner, eutectic-like composition in this figure, appear identical to those in the previous figure. It appears that poorly crystallized or amorphous SiC may exist in this sample as a dispersed phase in a Y-C-O matrix. This was the only reaction for which evidence of SiC was found, although that evidence is by no means conclusive. Predictions by Behrens and Brush (ref. 10) were based on the formation of binary phases only. The authors admitted that the existence of ternary and quaternary phases would indicate that their phase stability diagram "may not be entirely correct". The combustion temperature of this sample was not known, however at 1 atm pressure, Behrens and Brush predict YSi_2, YC_2, SiC and Y_2O_3 to form at temperatures below 2050 K. At 2050 to 2100 K, $YSi_{1.667}$, YSi_2, YC_2, SiC, and Y_2O_3 are predicted. From 2100 to 2500 K, $YSi_{1.667}$, YC_2, SiC, and Y_2O_3 are predicted. Only Y_2O_3 and YSi_2 were identified by powder diffraction data.

Reaction 2, the Zr-Si-C-O system, produced a specimen greatly different than did the first reaction. It produced the least dense, most crumbly specimen of the three proposed reactions. The polished specimen in figure 1 shows that only isolated areas densified. This suggests that this reaction was the weakest of the three with only a small amount of melting occurring. X-ray dot mapping was done on one of these isolated areas in figure 3. A BSE photo shows that two phases exist in the dense areas. The white phase may have a slightly higher concentration of Zr and Si than the dark phase. Beyond that, more information could not be obtained. Behrens and Brush predicted that between 1200 and 1500 K and for pressures between 10^{-4} and 10 atm, a relatively pure region of SiC and ZrO_2 would be found. No regions were predicted with Zr and C alone as identified by x-ray diffraction.

The last system studied, Ni-Zr-Si-C-O, like the first reaction, produced a specimen with dense regions which were surrounded by porosity (fig. 1). A low magnification (x500) BSE photo is shown in figure 4(a). The sample clearly contains at least three and possibly four or more phases. A light colored plate-like phase in a dark matrix as well as a second light colored phase which has a finer, more spheroid shaped morphology. Powder diffraction indicated that two or possibly three phases exist, ZrO_2, ZrC and possibly elemental Ni. However, elemental mapping did not support the existence of these phases. Oxygen and carbon were only found abundantly in the epoxy. The dark phase contains both Si and Ni. Zr is associated with the light colored phases as well as Si and Ni. At first glance, Si and Ni appear to be fairly evenly distributed, but on closer inspection, it can be seen that there is an area deficient in these elements that corresponds to the sphere shaped light colored phase dispersed around a circular crack in the sample. This is supported by figures 4(b) and (c) which were done at higher magnification. These showed that the light plate-like shaped phase contains Zr, Si and some Ni, although less than that in the dark phase. The light sphere shaped phase contains Zr, but no Si or Ni. It did show evidence of O and C however. The light phases may have produced the ZrC and ZrO_2 diffraction patterns. The dark phase is a Si-Ni material and is probably amorphous. Behrens and Brush did not model this reaction.

While these results were interesting, the attempt to fabricate a SiC based ceramic was not successful. It was expected that SiC would be formed with a second phase of Y_2O_3, ZrO_2 or ZrO_2 with Ni. However, the expected end products were not formed. The most promising reaction was the Y-Si-C-O system which may have produced some amorphous SiC. The modeling done by Behrens

and Brush did not successfully predict the phases found, particularly for the Zr-Si-C-O system. As the combustion temperature of the provided samples was not determined, it is possible that the temperature achieved during combustion did not correspond to the temperature necessary to synthesize SiC. Control of the temperature achieved during combustion is essential to controlling the end-products produced. However, as the modeling assumed only simple binaries were formed, it is likely that the stability diagrams are incorrect no matter what temperature was achieved during combustion.

ANALYSIS OF NICKEL ALUMINIDE SAMPLES

It was hoped that analysis of nickel aluminide samples which had been ignited from either the top or bottom would help determine if gravity had an effect on the SHS process. Nickel aluminides were chosen because melting occurs upon combustion in this system. The highest melting temperature for this system of compounds is for the equimolar compound NiAl, 1638 °C. As the adiabatic temperature is reported as 1650 °C (ref. 8), a liquid phase should occur with combustion. The presence of a liquid phase would increase the likelihood of gravity playing a role in this process via bulk flow. If microstructures of samples ignited from the top were different than those ignited from the bottom, it was felt that this would be evidence that gravity played a role. Further investigation would then be warranted. If no change in the microstructure was observed, then it was unlikely that gravity played a significant role in this process. Two samples each of four Ni-Al compositions were examined by x-ray powder diffraction, optical microscopy, scanning electron microscopy and electron microprobe. One sample was ignited so that the combustion wave traveled from the top to the bottom of the sample. The other was ignited at the bottom so combustion propagated from bottom to top. The expected reactions were:

(1) $Ni + Al \rightarrow NiAl$

(2) $2Ni + 3Al \rightarrow Ni_2Al_3$

(3) $3Ni + Al \rightarrow Ni_3Al$

(4) $Ni + 3Al \rightarrow NiAl_3$

Samples were evaluated at NASA following combustion at LANL. Each sample was sectioned through the center from top to bottom. Half of each specimen was mounted and polished and the other was ground to a powder and x-rayed. Table II shows the diffraction results for each reaction. The expected end-products were present. However, in these reactions as in the previous compositions examined, additional phases were present. No effect of the direction of the combustion wave was apparent from powder diffraction data obtained from powder taken from the top or bottom only of each specimen. In a couple of samples, slight differences in the intensity or type of phases present was observed, but further investigation would be necessary to determine if this was a result of combustion direction or simply a result of inhomogeneity in the sample. Initial analysis of the polished microstructures found no clear indication of gravity effects. Specimens of identical composition ignited in different directions had similar but not identical gross microstructures. An example is shown in figure 5. It was also found that microstructures can vary widely over a small area with abrupt transitions from one type of

microstructure to another. An example of this is shown in figure 6. This type of transition occurs throughout all of the samples. An example of variations in composition on a fine scale are shown in figure 7. Electron microprobe identified the phases in this nominally $NiAl_3$ sample as unreacted Al for the lightest phase, Ni_2Al_3 as the darkest phase and the medium gray phase as $NiAl_3$.

Continued examination revealed that specimens of different compositions, but with the same direction of combustion propagation, exhibited one thing in common. Samples which were ignited at the bottom so that the combustion wave propagated in an upwards direction showed a greater degree of pore coalescence than the samples ignited at the top. An example of this effect is shown in figure 8. No gradient in the pore size or pore number from top to bottom within each sample was found. At low magnification, the sample combusted from top to bottom does not exhibit the very large pores present in the sample combusted from bottom to top. Higher magnification photos show that on a fine scale, the sample combusted from bottom to top has much less porosity than the accompanying sample. However, careful analysis has shown that the overall porosity of the two specimens is the same. One expects porosity to occur with SHS processing as gases adsorped onto the starting powders surfaces must be released in someway. Also, pores naturally want to coalesce to cause a reduction in surface energy.

This apparent gravity effect supports the modeling results by Valone et al. (ref. 3) who found that gravity does not influence the kinetics of the combustion reaction, but predicted an effect of gravity on the resultant microstructure. Actual microstructural analysis is in agreement with this prediction. Specimens burned in an upward direction have a much coarser pore structure than those specimens burned in a downward direction. Thus one may reasonably expect that specimens produced in low gravity conditions would have finer pores than those produced on earth. But as overall percentage of porosity is not changed, the importance of this will depend on the application of the product.

EFFECT OF GREEN MICROSTRUCTURE

An interest in the effect of green microstructure on the final microstructure of combusted samples lead to development of the capability to combust samples at NASA Lewis. The microstructures of conventionally processed green bodies are known to have a large affect on their final microstructures (ref. 11). It was wondered if this was the case as well with SHS. Starting powders of Ti and C were either mixed by placing the powders in a 0.5 liter polyethylene mill with a half dozen milling media, but without milling fluid, or in a similar arrangement but with isopropyl alcohol as a milling fluid. The second method is a commonly used method of preparing powders for dry pressing (refs. 11 and 12). Both were milled for 24 hr. The slurry was dried in a pan on a hot plate prior to dry pressing. Cylindrical pellets were pressed and later combusted. It was expected that the specimens milled without a milling fluid present would exhibit a less uniform green microstructure than the other specimens and this was indeed the case. It was observed that some of the combusted specimens of TiC that had been made without milling fluid, showed evidence of pressing laminations persisting in the combusted microstructure (fig. 9). In some cases, these laminations halted the combustion front. This did not occur in samples that had been milled in a fluid. Work involving the introduction of seeded laminae confirmed this halt of the

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combustion front by laminations. Laminations were introduced by inserting thin pieces of plastic into the powder within a die prior to pressing a pellet. Location of the plastic was carefully noted for each sample. Laminations eliminated the contact area between planes of particles.

It was also found that a more uniform and reproducible microstructure was obtained by combusting samples within a spring loaded graphite die. Possibly this was because the graphite die retained much of the heat generated by combustion and thereby provided the samples with a more uniform thermal environment.

CONCLUDING REMARKS

The technique of self-propagating high-temperature synthesis was investigated to determine whether this technique fulfilled NASA needs for high temperature structural materials as well as for space and lunar materials. In one effort, it was found from microstructural and XRD analysis that initial attempts to use exothermic dopants to produce SiC based composites were not successful. It was concluded that simplified modeling by Behrens and Brush (ref. 10) did not correctly predict the phases present.

The second effort undertaken, to observe if there was an effect of gravity on the microstructures of combusted samples, was more successful. Larger pores were evident in samples which were ignited at the bottom so that the combustion wave propagated from the bottom to top of the sample, against gravity, than in samples ignited at the top so the combustion wave propagated from top to bottom. No variation in pore size or pore density was observed from bottom to top within any sample. These results indicate that gravity has an effect on pore size distribution, but not on overall porosity, phase composition or distribution. Modeling by Dr. Steven Valone at LANL (ref. 2) appear to indicate this same minor role for gravity's influence as well. These results suggest that a low gravity environment is desirable for SHS produced materials which melt during combustion.

In a third effort, green microstructure which is known to have a significant affect on structural ceramics, was also found to be important for certain SHS reactions as well. Fabrication laminations were found to impede the SHS reaction of Ti+C. However, improving green microstructure in Ti+C SHS samples was found to lead to a more uniform combusted microstructure. This study also showed that an improved microstructure can be obtained by enclosing samples in a graphite die during combustion in order to maintain a uniform temperature for a longer time than would be obtained without the die.

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TABLE I. – X-RAY RESULTS OF CERAMIC SYSTEMS

System	Proposed reaction	X-ray results
Y–Si–C–0	$4Y+3SiO_2+3C \rightarrow 2Y_2O_3+3SiC$	Y_2O_3 (major phase) YSi_2 (11–324) YSi_2 (11–596)
Zr–Si–C–0	$Zr+SiO_2+C \rightarrow ZrO_2+SiC$	Zr C
Ni–Zr–Si–C–0	$Si+2NiO+Zr+C \rightarrow SiC+ZrO_2+2Ni$	ZrO_2 (monoclinic) ZrC Unknown, low intensity $d \sim 2.04$ (Ni)

TABLE II. – X-RAY RESULTS ON Ni-Al COMPOSITIONS

Direction of combustion	Reaction	X-ray results
Top to bottom	$Ni+Al \rightarrow NiAl$	$NiAl$, Al_3Ni (minor phase)
Bottom to top	$Ni+Al \rightarrow NiAl$	$NiAl$, Al_3Ni (minor phase)
Top to bottom	$2Ni+3Al \rightarrow Ni_2Al_3$	$NiAl$, Al_3Ni (minor phase)
Bottom to top	$2Ni+3Al \rightarrow Ni_2Al_3$	Al_3Ni_2, Al_3Ni, Al (minor phase)
Top to bottom	$3Ni+Al \rightarrow Ni_3Al$	Ni_3Al
Bottom to top	$3Ni+Al \rightarrow Ni_3Al$	Ni_3Al
Top to bottom	$Ni+3Al \rightarrow NiAl_3$	$NiAl_3$, Al_3Ni_2, Al
Bottom to top	$Ni+3Al \rightarrow NiAl_3$	$NiAl_3$, Al_3Ni_2, Al
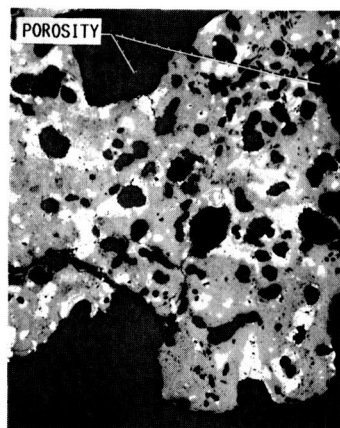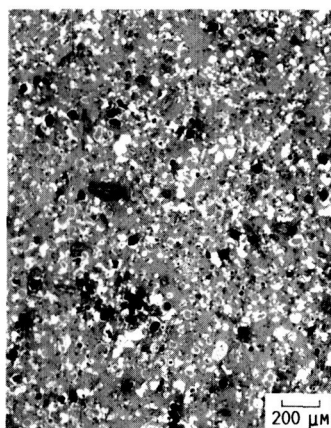
REACTION 1, $4Y+3SiO_2+3C$ REACTION 2, SiO_2+C+Zr GRAY IS POROSITY, WHITE IS DENSE PHASE REACTION 3, $Si+2NiO+Zr+C$

FIGURE 1. – OPTICAL MICROGRAPHS OF COMPOSITE MATERIALS FROM REACTIONS 1 THRU 3.

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BSE

YITTRIUM

SILICON

CARBON

OXYGEN

TA

20 μM
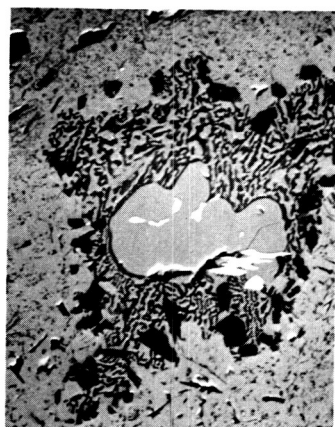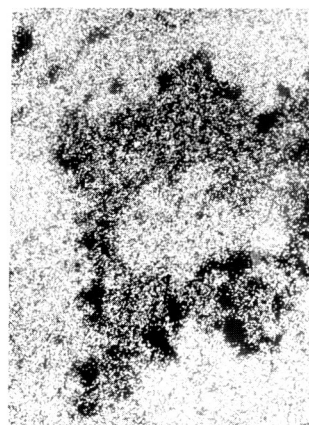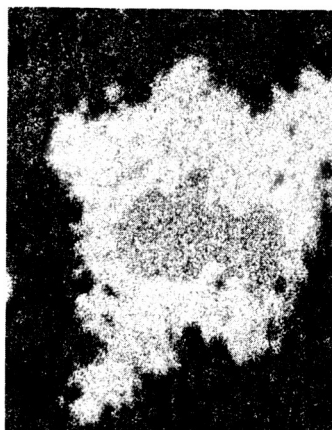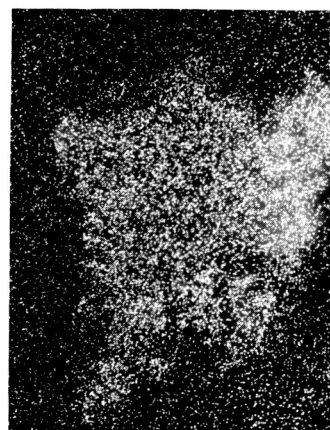
(A) SELECTED AREA FROM MICROSTRUCTURE OF REACTION 1.

FIGURE 2. – DOT MAPPING FROM REACTION 1, $4Y+3SiO_2+3C$.
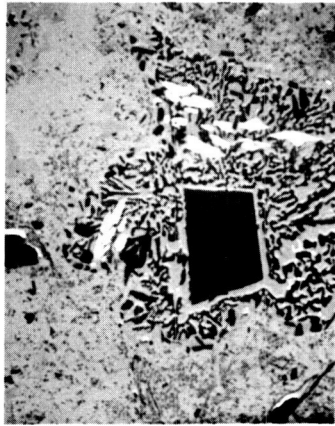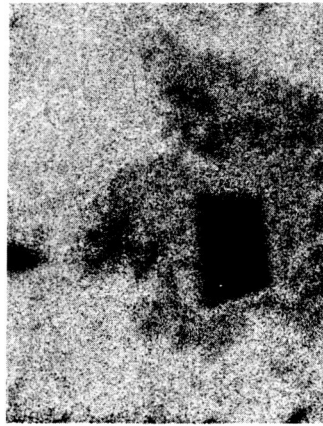
BSE

YITTRIUM

SILICON

CARBON

OXYGEN

TA

20 μM
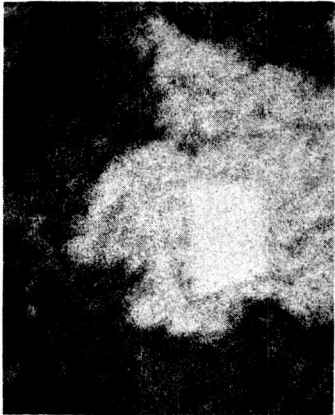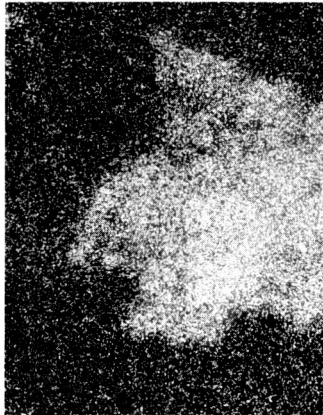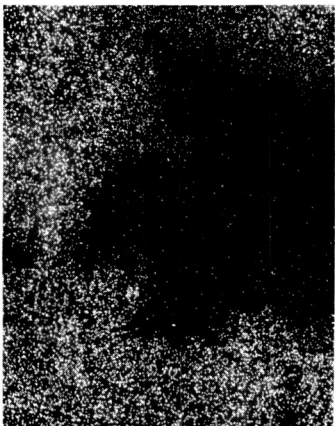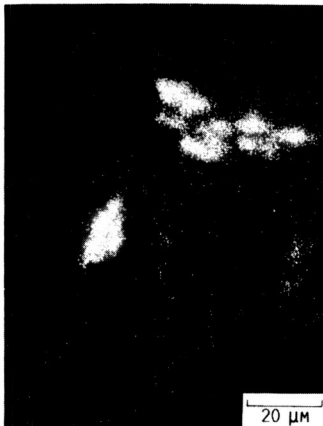
(B) SELECTED AREA FROM MICROSTRUCUTRE OF REACTION 1.
FIGURE 2. – CONCLUDED.

BSE

BSE

SILICON

CARBON

OXYGEN

Zr
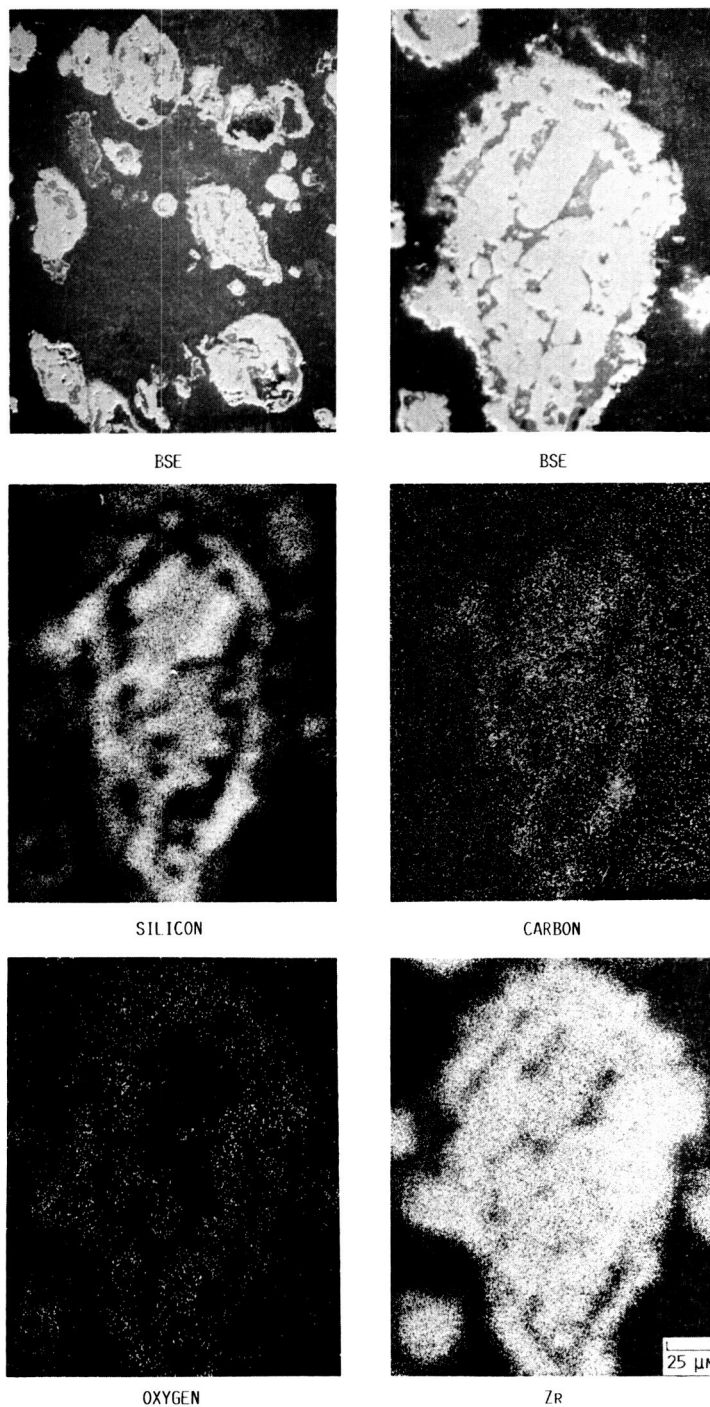
FIGURE 3. - DOT MAPPING FROM REACTION 2, $SiO_2 + C + Zr$.

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BSE

ZR

SILICON

CARBON

OXYGEN

NI

25 μM
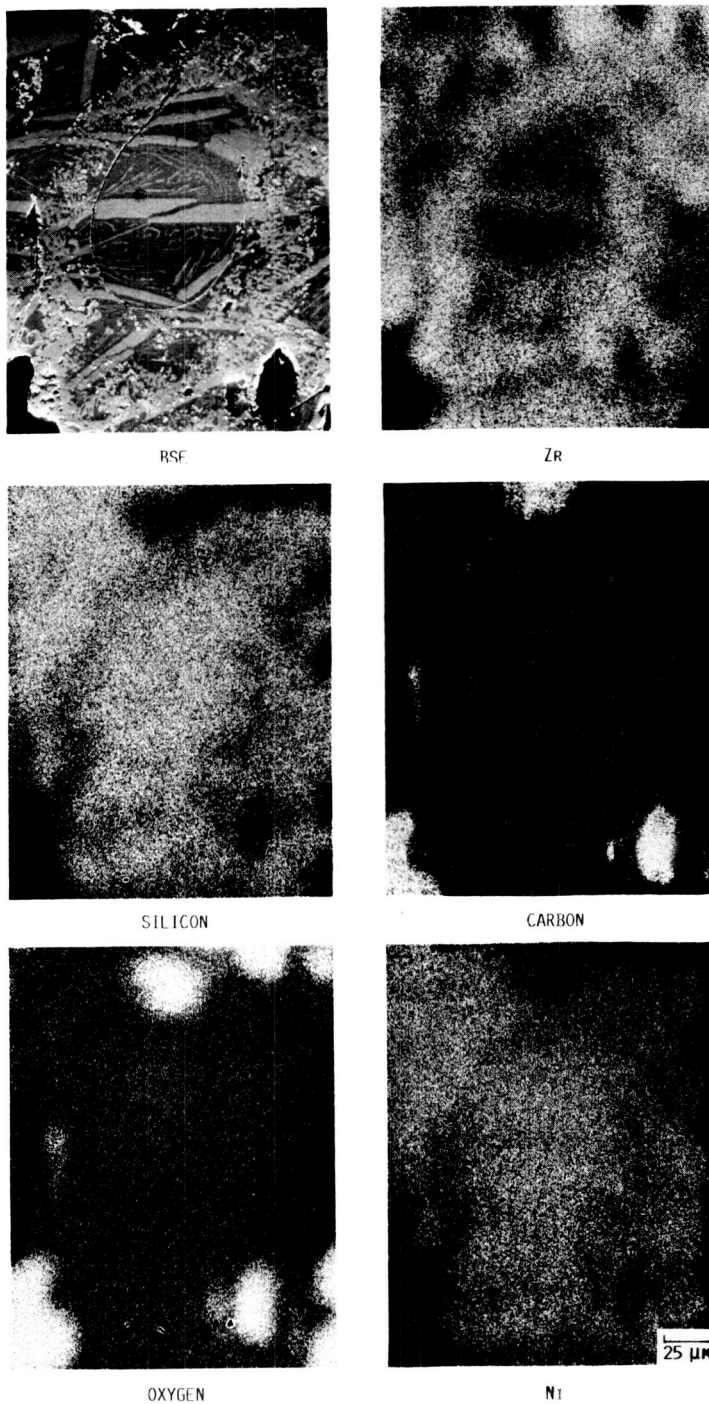
(A) SELECTED AREA FROM MICROSTRUCTURE OF REACTION 3.

FIGURE 4. - DOT MAPPING FROM REACTION 3, $Si+2NiO+Zr+C$.

BSE

ZR

SILICON

CARBON

OXYGEN

NI

10 μM
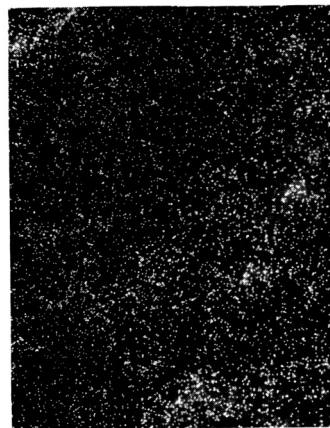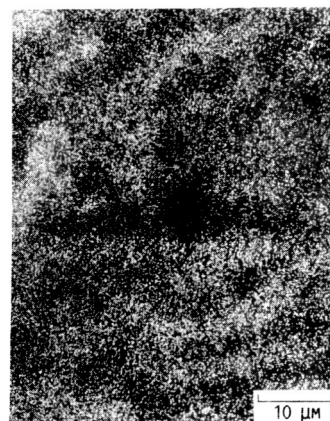
(B) SELECTED AREA FROM MICROSTRUCTURE OF REACTION 3.
FIGURE 4. - CONTINUED.

BSE

ZR

SILICON

CARBON

OXYGEN

NI

10 μM
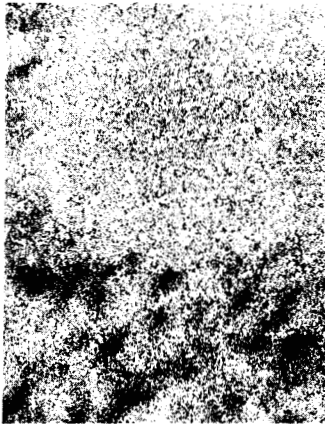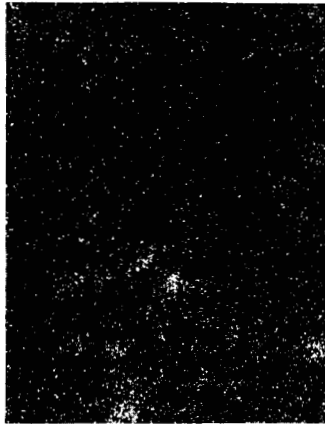
(C) SELECTED AREA FROM MICROSTRUCTURE OF REACTION 3.

FIGURE 4. - CONCLUDED.
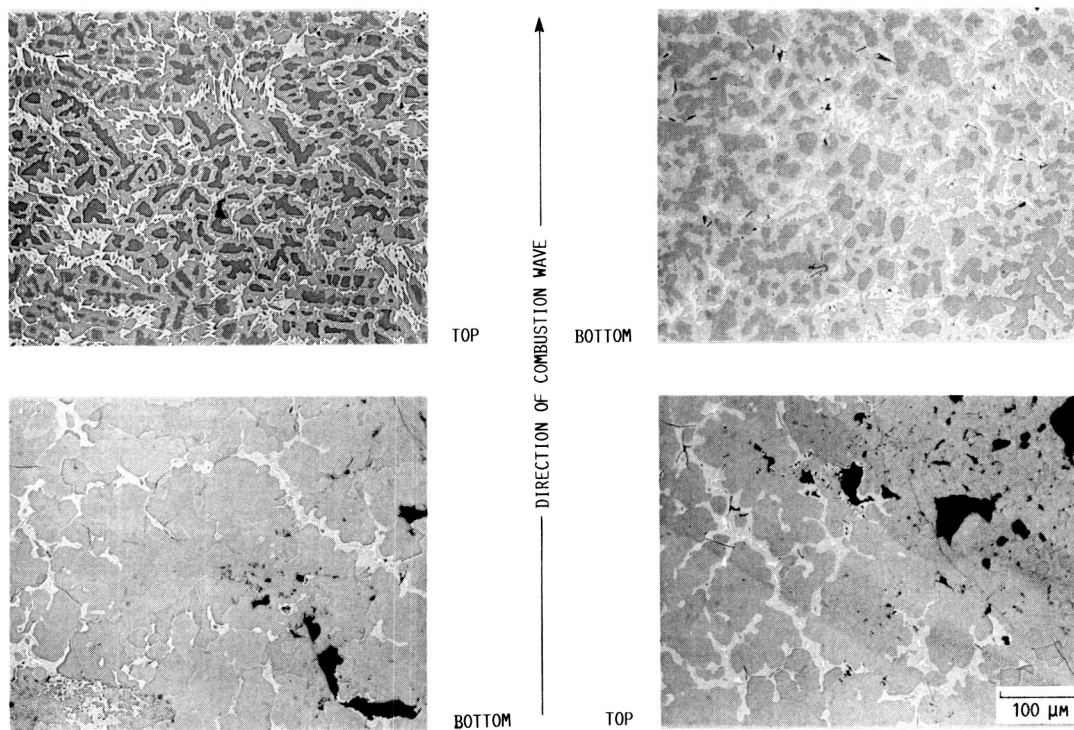
FIGURE 5. - COMPARISON OF MICROSTRUCTURES IN 2Ni+3Al SAMPLES IGNITED FROM DIFFERENT DIRECTIONS
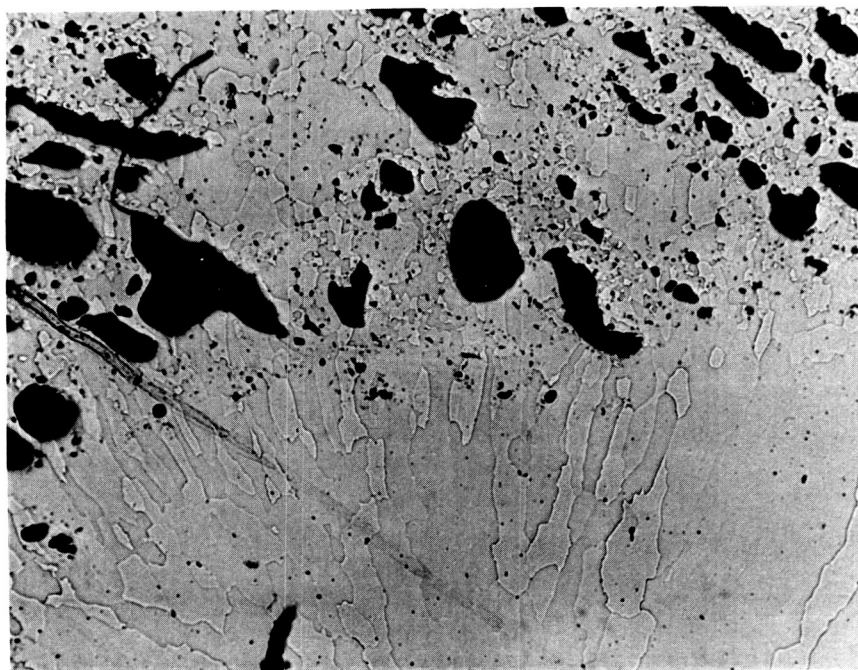
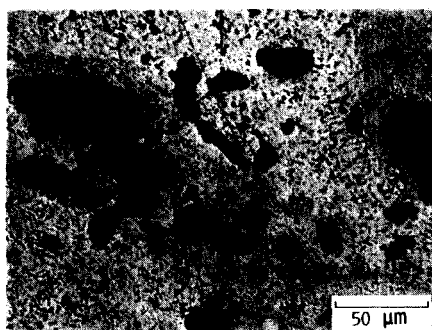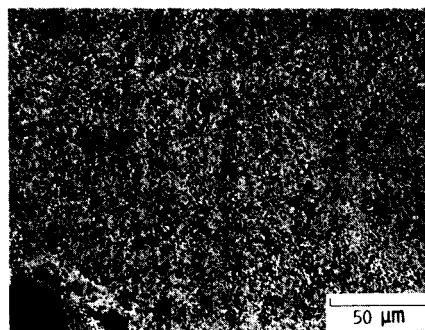FIGURE 6. - EXAMPLE OF ABRUPT TRANSITION IN MICROSTRUCTURE.

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FIGURE 7. - IDENTIFICATION OF PHASES IN A NiAl$_3$ SAMPLE. THE LIGHTEST PHASE IS Al, DARKEST PHASE IS Ni$_2$Al$_3$, THIRD PHASE IS NiAl$_3$.

COMBUSTED FROM BOTTOM TO TOP   COMBUSTED FROM TOP TO BOTTOM

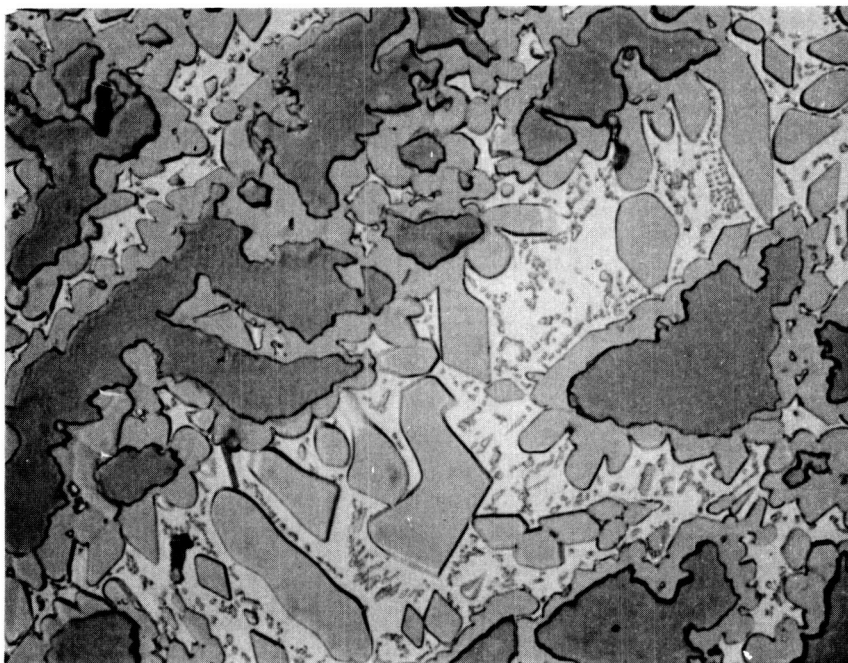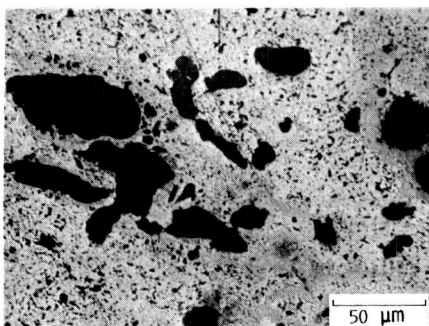FIGURE 8. - EXAMPLE OF THE AFFECT OF COMBUSTION DIRECTION ON PORE SIZE FOR REACTION 3 (Ni$_3$Al).

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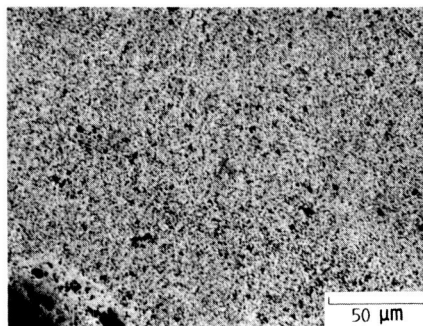
FIGURE 7. - IDENTIFICATION OF PHASES IN A NiAl$_3$ SAMPLE. THE LIGHTEST PHASE IS Al, DARKEST PHASE IS Ni$_2$Al$_3$, THIRD PHASE IS NiAl$_3$.

COMBUSTED FROM BOTTOM TO TOP

COMBUSTED FROM TOP TO BOTTOM

FIGURE 8. - EXAMPLE OF THE AFFECT OF COMBUSTION DIRECTION ON PORE SIZE FOR REACTION 3 (Ni$_3$Al).

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FIGURE 9. - PRESSING LAMINATIONS WHICH PERSIST IN THE COMBUSTED
MICROSTRUCTURE.

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15. Supplementary Notes

16. Abstract

This report summarizes three efforts aimed at investigating the process of self-propagating high temperature synthesis (SHS) for the fabrication of structural ceramics and intermetallics. Of special interest was the influence of processing variables such as exothermic dopants, gravity, and green state morphology in materials produced by SHS. In the first effort directed toward the fabrication of SiC, exothermic dopants of yttrium and zirconium were added to SiO_2 or $SiO_2 + NiO$ plus carbon powder mix and processed by SHS. This approach was determined to be unsuccessful since it did not produce the desired product of crystalline SiC. In the second effort, the influence of gravity was investigated by examining Ni-Al microstructures which were produced by SHS combustion waves traveling with and opposite the gravity direction. Although final composition and total porosities of the combusted Ni-Al compounds were found to be gravity independent, it was observed that larger pores were created in those specimens which were combusted opposite to the gravity force direction. Finally, it was found that green microstructure has a significant effect on the appearance of the combusted piece. Severe pressing laminations were observed to arrest the combustion front for TiC samples.

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